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Unraveling the size distributions of surface properties for purple soil and yellow soil

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ABSTRACT

Soils contain diverse colloidal particles whose properties are pertinent to ecological and human health, whereas few investigations systematically analyze the surface properties of these particles. The objective of this study was to elucidate the surface properties of particles within targeted size ranges (i.e. >10, 1–10, 0.5–1, 0.2–0.5 and <0.2 μm) for a purple soil (Entisol) and a yellow soil (Ultisol) using the combined determination method. The mineralogy of corresponding particle-size fractions was determined by X-ray diffraction. We found that up to 80% of the specific surface area and 85% of the surface charge of the entire soil came from colloidal-sized particles (<1 μm), and almost half of the specific surface area and surface charge came from the smallest particles (<0.2 μm). Vermiculite, illite, montmorillonite and mica dominated in the colloidal-sized particles, of which the smallest particles had the highest proportion of vermiculite and montmorillonite. For a given size fraction, the purple soil had a larger specific surface area, stronger electrostatic field, and higher surface charge than the yellow soil due to differences in mineralogy. Likewise, the differences in surface properties among the various particle-size fractions can also be ascribed to mineralogy. Our results indicated that soil surface properties were essentially determined by the colloidal-sized particles, and the <0.2 μm nanoparticles made the largest contribution to soil properties. The composition of clay minerals within the diverse particle-size fractions could fully explain the size distributions of surface properties.

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Introduction

Particle size distribution is a fundamental soil property that is commonly used to estimate other soil properties and processes, such as water retention characteristics (Guber et al., 2003; Henderson et al., 2005; Wösten et al., 2001), erosion capacity (Cheng et al., 2007; Guo et al., 2007; Wischmeier and Mannering, 1969), and transmission and distribution of nutrients and pollutants (Li and Pang, 2014; Six et al., 2002; Wang and Keller, 2008; Wang et al., 2006). Numerous studies

have demonstrated that soil surface properties profoundly influence these soil properties and processes (Bower and Gschwend, 1952; Ding et al., 2010; Fruhstorfer et al., 1993; Jenny and Reitemeier, 1934; Jiang et al., 2011; Li et al., 2010a; Petersen et al., 1996). Of course, the surface properties of soil and the particle size distribution should be interrelated.

Previous studies involving surface properties of soil particles in the sand (20–2000 μm), silt (2–20 μm) and clay (<2 μm) fractions have generally indicated that specific surface area (Dolinar, 2012; Ersahin et al., 2006; Petersen et

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al., 1996) and surface charge number (Ersahin et al., 2006; Yu, 1976) for a given soil are inversely related to particle size. In contrast to the substantial number of studies that have analyzed the surface properties of larger particles, studies on detailed size distributions of surface properties in the soil clay fraction ($<2 \mu\text{m}$) are rather scarce. Nevertheless, many soil processes are determined by the nature of the clay fraction (Hillel, 1998; Kiem and Kögel-Knabner, 2002; Saggari et al., 1999; Wu et al., 2012; Yonekura et al., 2013). In this size fraction, soil particles within the nanoscale or colloidal range ($<1 \mu\text{m}$) are vital and ubiquitous components in natural systems including soils, water, and atmospheric aerosol (Brunekreef and Russell, 2009; Schmidt, 2010). By virtue of their small size, high surface area and abundant surface defects, colloidal-sized particles are involved in the adsorption, migration and transformation of chemical contaminants, heavy metals, and nutrients (Grolimund et al., 1996; McCarthy and Zachara, 1989; Ryan and Elimelech, 1996; Wilkinson and Lead, 2007), which are key factors in several environmental problems (e.g., soil erosion, water eutrophication, heavy metal pollution and air pollution). Moreover, ingestion of fine-sized soil particles associated with many trace metals (Ajmone-Marsan et al., 2008; Madrid et al., 2008) and organic contaminants (Luo et al., 2011) through the water or air can even pose special risks to human health. Therefore, there is an increasing interest in extraction and characterization of colloidal-sized particles, especially natural nanoparticles in soils (Bakshi et al., 2014; Calabi-Floody et al., 2011; Li et al., 2010b; Li et al., 2012; Tian et al., 2014). For example, Calabi-Floody et al. (2011) extracted nanomaterials from the clay fraction of an Andisol and compared the zeta potential of these particles. However, little information on the systematic characterization of surface properties and mineralogy of soil colloidal particles in different size ranges was reported. Clearly, this information can provide insight into the interactions of colloidal-sized particles with environmental pollutants and ultimately clarify the role of colloidal particles in environmental problems.

Soils are composed of abundant clay minerals with constant-charge surfaces. Each kind of clay mineral has specific surface properties, and expansive clay minerals seem to have a dominant influence on soil properties due to their high specific surface area and surface reactivity. Given that the contents and proportions of mineralogical constituents are different in various soil fractions (Huang, 2000; Tsao et al., 2013), the mineral composition should be an important influence factor of the size distributions of soil surface properties. Nevertheless, the lack of the accurate determinations of different surface properties for soil particles within a targeted size range under the same experimental conditions handicaps the valid evaluation of their relationship with soil mineralogy, and ultimately hinders the quantitative assessment of their role in reactions and processes in soils. Fortunately, the combined determination method (Li et al., 2011) in this paper compensates for this shortage.

In this article, special attention was directed to yellow soil and purple soil, which are widespread in southwestern China. We systematically measured the surface properties and mineralogy of soil particles in different size ranges (i.e., >10 , $1-10$, $0.5-1$, $0.2-0.5$ and $<0.2 \mu\text{m}$) for these soils, with the aims

(1) to clarify the size distributions of surface properties for different kinds of soils, (2) to identify the composition of clay minerals in various particle-size fractions, and (3) to elucidate the relationship between surface properties and soil mineralogy in the size distribution.

1. Materials and methods

1.1. Separation and collection of soil particles within different size ranges for the tested soils

The yellow soil (Ultisol) and purple soil (Entisol) were collected from Chongqing, Southwestern China ($29^{\circ}45'N$, $106^{\circ}19'E$). Soil samples were taken from a depth of 0–20 cm within the profile, air dried at room temperature, gently crumbled and passed through a 2 mm mesh sieve to remove plant roots, stones and other large substances. A portion of each soil sample was used for basic chemical property analysis. The pH of the bulk soil suspension was measured at a soil/solution ratio of 1:2.5 in both ultrapure water and 1.0 mol/L KCl solution using a pH electrode. The organic matter content was measured by the potassium dichromate oxidation method (Yang et al., 2008). The cation exchange capacity was measured with the exchange method (Hu et al., 2000), and the free Fe oxide was determined by the citrate-bicarbonate-dithionite method (Lu, 2000). All the selected chemical properties of the bulk soils are displayed in Table 1.

Another portion of each soil sample was prepared for subsequent extraction of particles in different size fractions. Soil particles in size fractions of >10 , $1-10$, $0.5-1$, $0.2-0.5$ and $<0.2 \mu\text{m}$ were subsequently isolated according to the following procedure. First, air-dried soil particles for the purple soil and yellow soil were treated with 30% hydrogen peroxide (H_2O_2) to remove the associated organic matter (Calabi-Floody et al., 2011; House and Farr, 1989). The procedure involved ultrasonically dispersing the particles in ultrapure water, adding an appropriate amount of H_2O_2 solution to the suspension, heating at 60°C for 16 hr with intensive stirring, and washing away the excess H_2O_2 with ultrapure water. Then the obtained soil suspension was oven-dried at 75°C and passed through a $<0.25 \text{ mm}$ mesh sieve. Second, 50.0 g of the obtained soil particles was treated with 0.01 mol/L KOH solution in 500 mL of ultrapure water to adjust the pH value to 7.5. The suspension was then ultrasonically dispersed at a frequency of 20 kHz for 30 min by a probe-type ultrasonic homogenizer (Scientz-IID, Ningbo Scientz Biotechnology Co., Ltd., Zhejiang, China), before diluting to 5000 mL with ultrapure water. Finally, soil particles $<10 \mu\text{m}$ were extracted

Table 1 – Basic chemical properties of two tested soils.

| Soil types | pH | Organic matter content (g/kg) | CEC ($\text{cmol}_{(-)}/\text{kg}$) | Free iron oxides (g/kg) |
|-------------|-----|-------------------------------|---------------------------------------|-------------------------|
| Purple soil | 7.1 | 18.5 | 32.0 | 4.21 |
| Yellow soil | 4.4 | 2.85 | 11.9 | 10.4 |

CEC: cation exchange capacity.

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